



Europäisches Patentamt  
European Patent Office  
Office européen des brevets

⑪ Publication number:

0 339 111  
A1

⑫

## EUROPEAN PATENT APPLICATION

⑬ Application number: 88106777.1

⑬ Int. Cl.4: C08F 8/50 , C08F 114/26

⑭ Date of filing: 27.04.88

⑮ Date of publication of application:  
02.11.89 Bulletin 89/44

⑯ Designated Contracting States:  
CH DE FR GB LI

⑰ Applicant: SHAMROCK CHEMICAL  
CORPORATION  
Foot of Pacific Street  
Newark New Jersey 07114(US)

⑱ Inventor: Neuberg, William B  
Brittner Road  
Perrineville New Jersey 08535(US)  
Inventor: Luniewski, Robert  
14 Maureen Drive  
Smithtown New York 11788(US)

⑲ Representative: von Bezold, Dieter, Dr. et al  
Dr. Dieter von Bezold Dipl.-Ing. Peter Schütz  
Dipl.-Ing. Wolfgang Heusler Briener Strasse  
52  
D-8000 München 2(DE)

### ⑳ Apparatus and method for radiation processing of materials.

㉑ A method and apparatus for radiation degradation processing of polytetrafluoroethylene makes use of a simultaneous irradiation, agitation and cooling. The apparatus is designed to make efficient use of radiation in the processing.

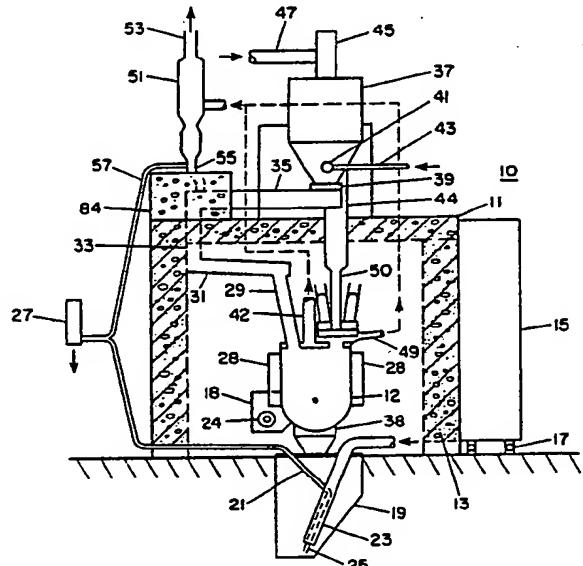


FIG. 1

EP 0 339 111 A1

## Apparatus and Method For Radiation Processing of Materials

### Background of the Invention

This invention relates to radiation processing for the degradation of materials and specifically to radiation degradation of polytetrafluoroethylene (PTFE).

U.S. Patent 3,766,031 to Dillon discloses a method for radiation processing of polytetrafluoroethylene, wherein the polytetrafluoroethylene is exposed to radiation and thereafter subjected to comminution to reduce the polytetrafluoroethylene to a fine particle powder. This powder is useful as a dry lubricant, for example, in paints and inks. This and other methods for carrying out this technique generally made use of electron beam or cobalt sources for irradiating the polytetrafluoroethylene. The material being irradiated was arranged in trays and exposed to multiple doses of radiation, approximately 2 to 15 MR per pass, so that the temperature of the material does not rise excessively, thereby to avoid discoloration of the material and the possible generation of noxious gases. The total dose is approximately 35 to 150 MR.

The tray irradiation technique has a radiation utilization efficiency of only approximately 35%. Radiation inefficiencies arise from the three factors, which are (1) overscan of trays, (2) gaps between trays and (3) depth-dose characteristics for an electron beam.

The need to assure complete and uniform radiation of a tray requires some overscan by the radiation and an efficiency loss of 5 to 15%. Likewise there is usually some space between trays of material which causes a further 10 to 15% loss of efficiency.

The largest efficiency loss arises out of the fact that the dose received by the material varies with material depth. This variation is illustrated in the graph of Figure 6. Typically the dose at the surface, designated R, is taken as the nominal dose for the material. Beam energy and/or material depth is adjusted so that an equal dose at the opposite surface of the material (Depth D). Radiation which passes entirely through the product (Area C) is not utilized. Radiation in Area B in excess of the nominal dose R, is likewise not used. This causes further inefficiency, and in some instances may result in undesired properties of the resultant product. Overall this depth-dose characteristic can cause processing inefficiency of up to 50%.

The prior art technique of slow irradiation of material by subsequent exposures to doses of radiation, occasionally coupled with stirring the material

between exposures, tends to cause the production of radiation degraded polytetrafluoroethylene powder to be an expensive, inefficient and time consuming operation.

It is an object of the present invention to provide an apparatus and method for more efficiently producing radiation degraded polytetrafluoroethylene.

### Summary of the Invention

In accordance with the present invention there is provided an apparatus for radiation processing of chopped flowable solid materials having a high molecular weight to degrade the material to a lower molecular weight and render it grindable into a powder. The apparatus includes a processing vessel for holding the material, a radiation source for supplying radiation to the processing vessel, means for agitating the material during the processing, and cooling means for maintaining the vessel and the material below a selected temperature.

In a preferred embodiment the apparatus includes a dust cover over the vessel and dust collecting means for gathering airborne material particles which accumulate in the dust cover. In connection with the processing, air or water may be supplied to the material in order to fluidize the material, and to additionally cool the material by the flow of air or the addition of cooling water.

The processing vessel is preferably a ribbon blender which includes a vessel and motor driven stirrer for agitating the contents of the vessel. A preferred radiation source is a source of high energy electrons. The processing vessel can be cooled by a water flow channel surrounding the vessel. Cooling can also be accomplished, alternatively in part, by the addition of water to the processing vessel.

In accordance with the invention there is provided a method for processing flowable solid polytetrafluoroethylene material by radiation degradation to reduce the molecular weight of the material and render it grindable into a powder. The method includes exposing the material to radiation, agitating the material during exposure, and maintaining the material at a temperature below 500°F during exposure.

In preferred embodiments the material is fluidized by an air flow during exposure and maintained below 250°F or, alternatively, water is added to the material during exposure and the mixture maintained within an appropriate temperature

range.

For better understanding of the present invention, together with other and further objects, reference is made to the following description, taken in conjunction with the accompanying drawings, and its scope will be pointed out in the appended claims.

#### Brief Description of the Drawings

Figure 1 is an end elevation view of a material processing apparatus in accordance with the present invention;

Figure 2 is a partial cross-sectional view of the Figure 1 apparatus showing an air supply manifold;

Figure 3 is a cross-sectional view of a portion of the electron beam radiation source of the Figure 1 apparatus;

Figure 4 is a cross-sectional view of the processing vessel of the Figure 1 apparatus;

Figure 5 is a side view of the processing vessel of the Figure 1 apparatus;

Figure 6 is a graph illustrating the dose of radiation received by material to be processed at various depths;

Figure 7 is a cross-sectional view, taken along the view line VII-VII of Figure 8, of a processing vessel similar to that shown in Figure 1 but with certain alterations; and

Figure 8 is a side view of the processing vessel shown in Figure 7.

#### Description of the Invention

Figure 1 is an end elevation view of an apparatus 10 for processing material in accordance with the present invention. Apparatus 10 is particularly useful for performing radiation degradation of polytetrafluoroethylene in order to degrade the material by reducing its molecular weight and render the material grindable into a powder.

Apparatus 10 includes a processing vessel 12, which comprises the vessel of a ribbon blender, shown in greater detail in Figures 4 and 5. Vessel 12 includes a longitudinal shaft 14 having attached thereto ribbon blending paddles 16. Processing vessel 12 and its associated equipment are enclosed in a radiation chamber 11, made of masonry or other radiation attenuating material. Chamber 11 has an access opening 13 which is closed by door 15 mounted on rollers 17. Door 15 is provided for equipment access, and other openings, such as a zig-zag hallway, may be provided for personal access. As may be more easily seen by reference to Figures 4 and 5, the paddles are arranged so that

the material within the ribbon blender is moved axially back and forth in the longitudinal direction of shaft 14 when the shaft is continuously rotated in the direction of arrow 59, and therefore the material undergoes no average axial displacement within the vessel.

Shaft 14 is driven by reduction gear 18 which is driven by motor 24. In a typical embodiment the shaft rotates at a speed of approximately 40 revolutions per minute, stirring and agitating material 60 which has been placed on the inside of the vessel 12.

In one embodiment, the vessel 12 has paddles with a radius of 18 inches and is 72 inches in length. The vessel is provided with a remote control outlet opening 38 in the bottom, for use in draining material after processing. Opening 38 is arranged above material receiving chamber 19, so that upon opening processed material in vessel 12 will flow into the receiving chamber 19. The material is removed from the receiving chamber by a vacuum apparatus 27 which draws the material into nozzle 25 and through conduit 21. Air inlet conduit 23 is arranged concentric to nozzle 25 to provide an air supply for material removal by the vacuum apparatus. Alternately a screw conveyor might be used.

In addition, the vessel is provided with a cooling jacket 28 for the flow of cooling water in order to maintain the vessel and its contents at a temperature below approximately 500°F, preferably below approximately 250°F. Water is provided through inlet and outlet conduits 30 and 32 and flows between separate portions of jacket 28 via conduit 61.

The cooling water is preferably supplied at a temperature which is above the dew point of the surrounding air to avoid condensation. Because the radiation processing of PTFE can release fluorine gas, which combines with water to form hydrofluoric acid, such condensation can cause acid formation and rapid corrosion of the equipment. One method of increasing supply water temperature is to use the cooling water to first cool the power supply of the electron beam apparatus, and thereafter use the same water at elevated temperature to cool the vessel. Instead of using the same water, a water-to-water heat exchanger can be used. Another technique is to use a conventional water heater to maintain water temperature above the dew point.

The apparatus of the present invention also includes mechanisms for supplying flowable solid material to the processing vessel 12. This equipment includes a generally vertical chute 29 leading into the processing vessel and communicating at its upper end with a horizontal screw conveyor 31, which is located within radiation chamber 11. Verti-

cal chute 33 passes through the top of radiation chamber 11 and connects interior screw conveyer 31 with exterior screw conveyer 35. The region surrounding vertical shaft 33 is provided with additional radiation shielding structure 84, which may be fabricated out of bricks or the like. Exterior screw conveyer 35 receives granular material to be processed from the bottom of hopper 37, which is provided with a remote control valve 39, and a vibrator 41 operated by compressed air supplied over conduit 43. Granular material is supplied to hopper 37 by conduit 47 through which material is drawn by vacuum apparatus 45.

Processing vessel 12 is provided with a cover 40 and a vent conduit 42 through which air is exhausted under lower than atmospheric pressure in order to remove airborne material particles and fluorine gases from the processing vessel so that they do not accumulate and corrode the window of the electron beam source. An additional venting conduit 49, having lower end 82 with a nozzle 84, arranged near the outlet of the electron beam source, may also be provided.

Conduits 42 and 49 are connected to a cyclone separator 51, which separates airborne material particles from exhaust gases, which are vented through conduit 53. Particles which accumulate in separator 51 may be returned to the processing vessel over conduit 55 during initial stages of the processing. At later stages of the processing, the material collected by separator 51 is sufficiently degraded so that it may be provided over conduit 57 to output vacuum apparatus 27.

The electron beam accelerator, which is used to provide a radiation source for the Figure 1 apparatus includes a vacuum output passage 44 which is connected to a sectoral horn 50 by a beam steering section 46 (not shown in Fig. 1), which is provided with an electro-magnet 48, shown in Figure 5. The electron beam source used has an electron acceleration of 1 million volts, and can provide beam current of up to 100 milli-amperes. Beam steering section 46 causes the beam to oscillate back and forth in the sectoral horn 50 to assume beam paths 52, 54 and 56, illustrated in Figure 5, thus spreading the radiation along the length of vessel 12. The electron beam exists from the vacuum portion of sectoral horn 50 through primary window 58 and optional secondary window 66, respectively, which are shown in detail in Figure 3. The space 68 between the primary and secondary windows is provided with a flow of cooling air through conduits 70 and 72. In addition, conduits 74 and 76 are provided on flange 62, and possibly also on flange 64, to provide water cooling of the windows. Cooling water is supplied through conduits 78 and 80 and preferably has a temperature above the dew point of the surrounding air to

avoid condensation and possible formation of hydrofluoric acid as described above. Windows 58 and 66 comprise thin sheets of titanium, with a thickness of between 0.00075 inch and 0.002 inch.

5 Primary window 58 has a preferred thickness of 0.0015 inch in order to support vacuum pressure. Secondary window 66 does not require the same mechanical strength, but a comparable thickness is preferred in order to provide corrosion resistance.

10 In connection with the processing of materials, such as PTFE in accordance with the present invention it is desirable to supply a flow of air to the material in the processing apparatus. The air flow serves three functions: (1) the supply of air tends to provide fluidizing of the flowable solid material so that it flows easily when agitated by stirrer paddles 16; (2) the supply of air tends to promote the degradation reaction, which requires oxygen; (3) the supply of air into the reaction vessel and out 15 through conduit 42 tends to promote cooling of the material, as a supplement to the cooling provided by water jacket 28.

20 In the embodiment shown in detail in Figure 2, air is provided to the reaction vessel through manifold 34, which has air outlet passages 35 directed into the material contained in the vessel. Compressed air is supplied over conduit 36.

25 In a typical process, flowable solid material such as virgin or scrap polytetrafluoroethylene, in 30 course powder or chip form is placed into the vessel 12 and subjected to agitation by the rotation of stirrers 16 while undergoing irradiation by means of electron beams supplied from electron source 44. The electron beam from source 44 is steered in 35 vacuum passage 46 by alternating currents applied to electro-magnet 48 so that the electron beam diverges in sectoral horn 50 into paths which are spread out in one angular direction along the length of the vessel 12 as indicated by beams 52, 54 and 40 56. The action of stirrers 16 rotating in direction 59 tends to circulate the material for uniform irradiation. The electron beams exit from the sectoral horn vacuum through windows 58 and 66. The 45 irradiation by the electron beam, in combination with the stirring action of stirrer 16 of the ribbon blender causes the degradation of the material to lower its molecular weight from around 5,000,000 to 10,000,000 to less than 1,000,000 and probably less than 100,000. When the molecular weight of 50 the material is thus lowered, it becomes grindable into a powder. In order to promote the reaction by supply of oxygen, provide further cooling of the material during the irradiation process, while the material is undergoing reaction, and in order to 55 make the material more fluid and easier to stir, air may be provided by conduit 36 into manifold 34 which has outputs 35 distributed along the length of vessel 12.

By reason of the agitating action of the stirrers 16, in addition to the air provided to conduit 34, the process may produce a considerable amount of airborne particles. These particles are constrained within dust cover 40, which is provided with an outlet conduit 42, which is positively ventilated through dust collector 51, whereby the recovered material can be recycled into the process vessel or collected as degraded, low molecular weight product through conduits 55 and 57.

It should be noted as shown in the cross-sectional view of Figure 4 that the action of the stirrers 16 rotating in the direction of arrow 59 will cause the process material 60 to build up on one side, for example, the right side, of the reaction vessel 12. In order to provide the most effective radiation treatment, the sectoral horn 50 is directed to a selected region of irradiation at this side of the reaction vessel, so that the electron beams intercept the maximum amount of material to be processed. By directing radiation into the portion of the vessel having a larger depth of material, the efficiency of the process is enhanced, since essentially all of the incident radiation is intercepted by material, rather than passing through without reaction. Further, overradiation which might normally occur for material close to the surface upon which the radiation is incident is avoided by reason of the agitation of the material by the paddles 16.

In order to provide processing of polytetrafluoroethylene in accordance with the method of the present invention using the apparatus of Figure 1, scrap or virgin polytetrafluoroethylene in flowable solid form such as coarse powder or chips, either sintered or unsintered, is supplied to the vessel through chute 29. Typically, during processing the volume of the charge of material provided to the reaction vessel is reduced as the bulk density of the material is increased during processing. Processing of a batch of material (approximately 3200 lbs.) may use an electron beam accelerated at 1 million volts with a current of 20 to 100 millamps for six or seven hours. As an alternate to the use of an electron beam accelerator, a radioactive material, such as Cobalt-60, can provide the radiation. Typically, only a portion of the full load of material is initially supplied to vessel 12, for example, one-third to one half the load. Since the bulk density of chip form PTFE is generally low, this fills the volume of the vessel. Following initial irradiation, additional material can be added because of the increase in bulk density and consequent reduction of material volume during irradiation. After the full load of material is provided, radiation, agitation and cooling continues until the material receives a dose of 125 to 150 Mrad per pound. During this processing, the temperature of the material is monitored by thermal

5 sensor 88, which is connected by wire 89 to remote meter 90. When the temperature of the material exceeds a selected value, the radiation beam current is reduced. The maximum temperature should be below 500° F, preferably below 250° F.

10 It has been found that the polytetrafluoroethylene can be cooled and an improved product obtained by supplying water to the processing vessel 12 during processing. Referring to Figures 7 a,d 8 which show processing vessel 12, a pipe 91 supplies cooling water to a manifold 92 from which extend tubes 93. This arrangement facilitates the addition of water to the material 60 being processed in the vessel 12. In this preferred embodiment, the cooling jackets 28 shown in Figure 4 have been removed and cooling jackets 94 supplied with cooling water through inlet and outlet conduits 95 and 96 have been added to the ends of the vessel 12. Water flows between the jackets through a conduit, not shown, joining them together. In addition, conduits 97 and 98 have been affixed to the edges surrounding the opening in cover 40 to provide water cooling of the cover to prevent warping due to the high temperatures otherwise encountered. As explained heretofore, all of the cooling water supplied has a temperature within the dew point of the surrounding air to avoid condensation and the formation of hydrofluoric acid.

15 30 Instead of using the conduit 91 and manifold 92 to supply water to the vessel, a manifold can be positioned above the vessel 12 and four tubes extended downwardly therefrom through the cover 40 to supply water to the material being processed. The manifold above the vessel is fed cooling water through a suitable inlet conduit.

20 35 40 It was anticipated that the addition of water to the material being processed would provide cooling by evaporation. It was discovered, however, that the initial amounts of water added to the polytetrafluoroethylene caused an increase in temperature of the material, contrary to expectations. While the reasons for the temperature increase are not completely understood, it is believed that the water undergoes a reaction with the polytetrafluoroethylene. It is suspected that this reaction includes the combining of water with free fluorine in the vessel to form hydrofluoric acid.

45 50 55 The temperature increase caused by the reaction of water with the material is greatest when the material being processed is that known as virgin polytetrafluoroethylene. When that material is being processed, it has been found that water should be added to the vessel when the material reaches a temperature of about 260° F. With a batch of polytetrafluoroethylene of about 3000 pounds in the vessel, a water flow of 1/2 liter to 2 liters per minute has been found satisfactory. When the temperature of the processed material is lowered to

about 245° F, the water flow is diminished or interrupted until the temperature again increases to about 260° F, at which time water is again supplied to the vessel.

Experiments have shown that if the temperature of virgin polytetrafluoroethylene is permitted to rise as high as 265° F, the addition of water will cause the temperature to increase further, due to the reaction of water with the materials in the vessel, and it then becomes necessary to interrupt the processing to cool the mixture and maintain it in a stable condition, thereby preventing discoloration.

When the polytetrafluoroethylene processed is what is called "hard stock," i.e., sintered scrap polytetrafluoroethylene which has already had a heat treating history, the increase in temperature caused by the addition of water is less than with the virgin polytetrafluoroethylene. In processing this material, it has been found that its temperature can be allowed to rise to about 390° F, at which point water is added and the material cooled to about 375° F before the water supply is diminished or interrupted.

As a further example, when processing the materials known as (1) granular polytetrafluoroethylene (also known as "white tails"), (2) scrapped teflon tape or (3) non-virgin polytetrafluoroethylene, or a combination of these materials, the temperature can be allowed to rise to about 265° F before water is supplied to the material. After cooling to about 250° F the water supply is diminished or interrupted until the temperature again rises to about 265° F.

It has been unexpectedly found that the addition of water to the process material causes an improvement in the oil absorption characteristics of the resulting powder, i.e., its oil absorption characteristic is lowered. A powder with lower oil absorption when mixed in a carrier such as an oil or solvent for paint, provides a flowable consistency like honey with low thickening properties. This is generally desirable. A material with higher oil absorption when mixed with a carrier, provides a consistency more like peanut butter, i.e., higher thickening properties, and is less desirable as a product. The material produced when cooling water is added to the polytetrafluoroethylene during processing has the desirable low oil absorption property, and this is a superior product for many applications.

While there has been described what is believed to be the preferred embodiment of the present invention, those skilled in the art will recognize that other and further modifications may be made thereto without departing from the spirit of

the invention, and it is intended to claim all such embodiments as fall within the true scope of the invention.

5

### Claims

1. Apparatus for radiation processing of high molecular weight, flowable solid polytetrafluoroethylene material to degrade said material to lower its molecular weight and render it grindable into a powder, including a processing vessel for holding a batch of said material during processing, a source for supplying radiation to said processing vessel, characterized in that said radiation is provided to a selected region of said processing vessel; that said processing vessel is provided with mechanical stirring means for agitating said material in said processing vessel during said processing thereby to repeatedly move said material into and out of said selected region whereby said material is uniformly irradiated and in that there is provided cooling means for maintaining said vessel and said material below a selected temperature during said processing:
2. Apparatus according to claim 1 further characterized in having a dust cover over said vessel and dust collection means associated with said dust cover.
3. Apparatus according to claim 1 or claim 2 further characterized in means for supplying air to said material thereby to cool said material, to maintain said material in a fluidized state and to provide oxygen to said material.
4. Apparatus according to any of claims 1 through 3 further characterized in a radiation shield surrounding said processing vessel and said radiation source and means for supplying said material through said radiation shield to said vessel.
5. Apparatus according to any of claims 1 to 4 further characterized in that said vessel comprises a ribbon blender having a motor driven stirrer and includes a water jacket for cooling said vessel and said material.
6. Apparatus according to any of claims 1 to 5 further characterized in that said radiation source comprises a source of high energy electrons and includes deflector means for distributing said electrons over said selected region.
7. Apparatus according to claim 6 wherein said radiation source is further characterized by having an aperture with primary and secondary aperture windows and wherein there is provided flow of air between said windows.
8. Apparatus according to claim 6 wherein said radiation source is further characterized in having an aperture window and means for water cooling

said aperture window with cooling water at a temperature above the dew point of the surrounding air.

9. Apparatus as specified in any of claims 1 through 8 further characterized in that said cooling means comprises means for supplying water to said material.

10. A method for processing flowable solid polytetrafluoroethylene material by radiation to degrade said material to lower its molecular weight, wherein said material is supplied to a processing vessel and irradiated in the processing vessel characterized in that said radiation is supplied to a selected region of said processing vessel, said material is agitated in the processing vessel during said radiation thereby to repeatedly move said material into and out of said selected region so that said material is uniformly irradiated and wherein the material is cooled to maintain a selected temperature below 500 °F during said processing.

11. A method according to claim 10 further characterized in that air is provided to said material to cool said material, fluidize said material and provide oxygen to said material during said radiation.

12. A method according to claim 10 or 11 further characterized in that additional material is supplied to said vessel during said processing.

13. A method according to any of claims 10 through 12 further characterized in that air is supplied to said material during said processing and that said vessel is vented by negative pressure to remove airborne particles and gases.

14. A method according to claim 13 further characterized in that said removed material is recovered.

15. A method according to any of claims 10 to 14 further characterized in that water is supplied to said material during said processing.

16. A method according to any of claims 10 to 15 wherein the material is virgin polytetrafluoroethylene, further characterized in that the selected temperature is about 260 °F.

17. A method according to claim 16 further characterized in that said cooling is interrupted when the temperature of the material is lowered to about 245 °F.

18. A method according to any of claims 10 to 16 wherein the material processed is sintered scrap polytetrafluoroethylene which has a heat treatment history, further characterized in that said selected temperature is below about 390 °F.

19. A method according to claim 18 further characterized in that cooling is interrupted when the temperature is lowered to about 375 °F.

20. A method according to any of claims 10 to 16 wherein the material processed is non-virgin polytetrafluoroethylene, further characterized in that said selected temperature is about 265 °F.

21. A method according to claim 20 further characterized in that cooling is interrupted when the temperature of the material is lowered to about 250 °F.

22. A method according to any of claims 10 to 16 wherein the material is granular polytetrafluoroethylene chips further characterized in that said selected temperature is about 265 °F.

23. A method according to claim 22 further characterized in that cooling is interrupted when the temperature of the material is lowered to about 250 °F.

24. A method according to any of claims 10 to 16 wherein the material processed is scrap polytetrafluoroethylene tape, further characterized in that said selected temperature is about 265 °F.

25. A method according to claim 24 further characterized in that cooling is interrupted when the temperature is lowered to about 250 °F.

5

10

15

20

25

30

35

40

45

50

55

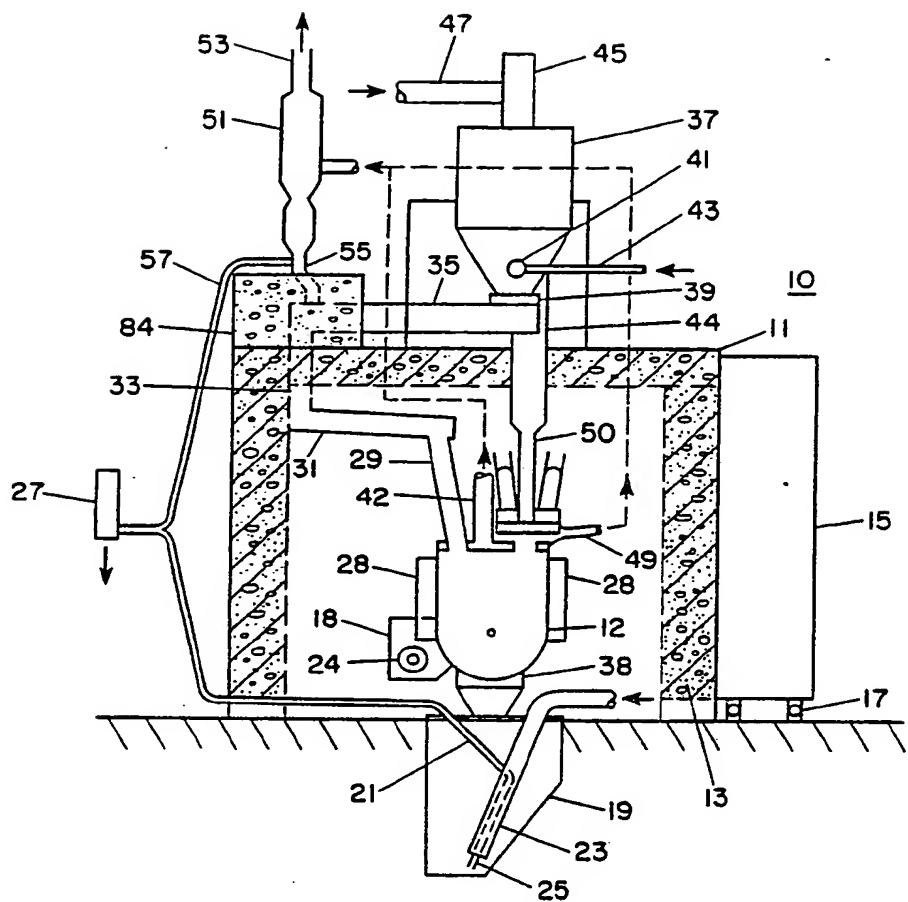


FIG. 1

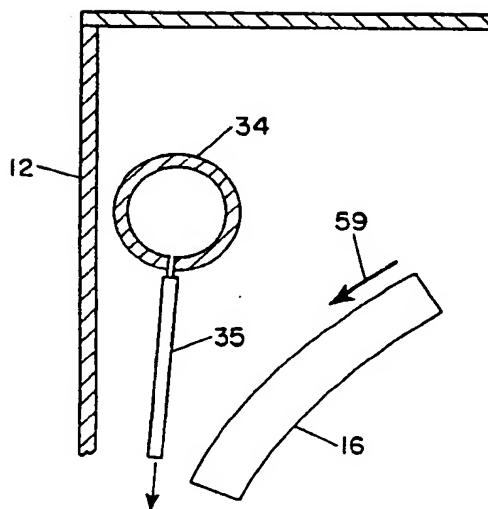


FIG. 2

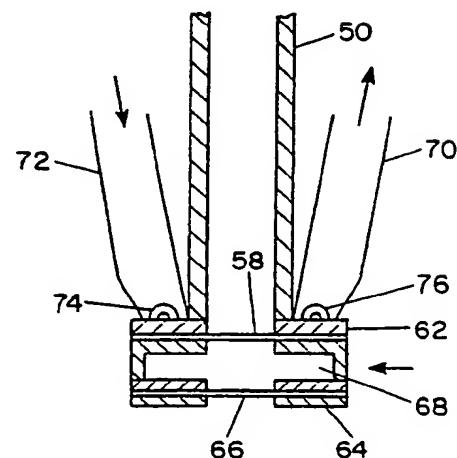


FIG. 3

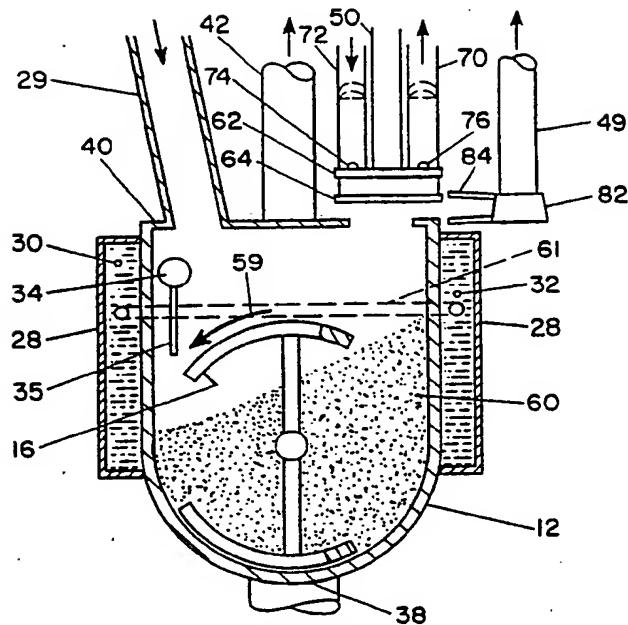


FIG. 4

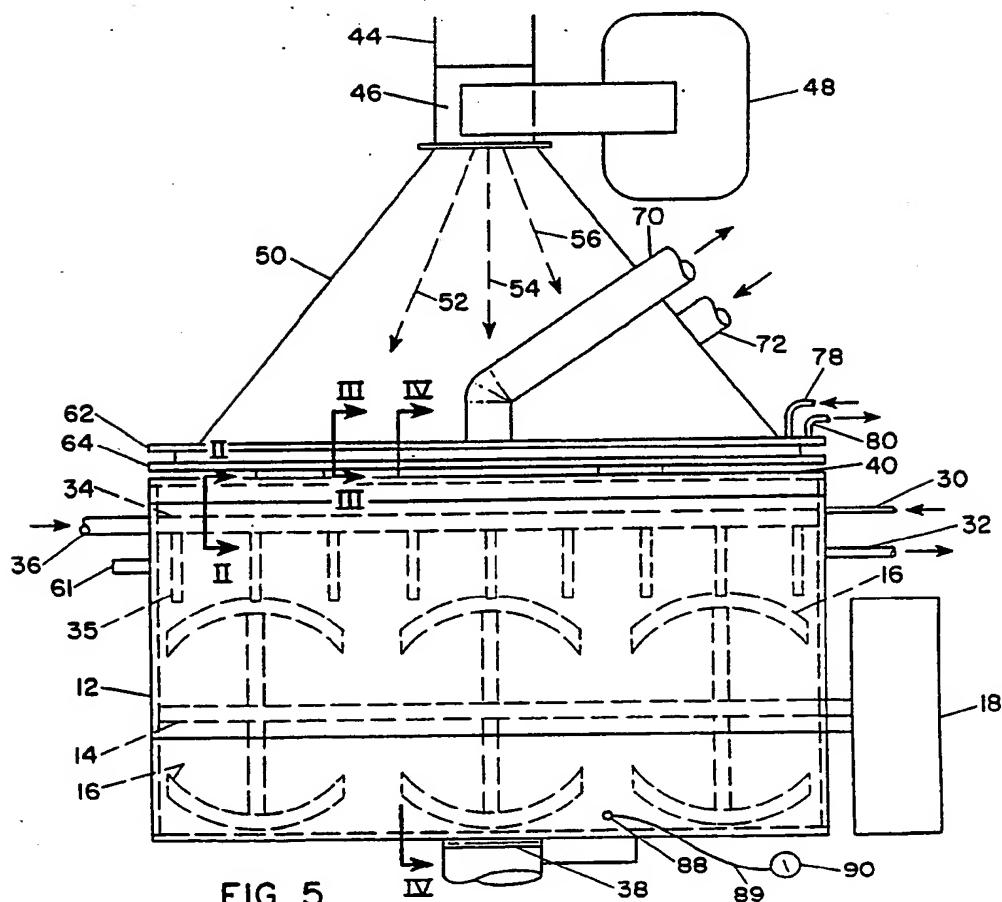


FIG. 5

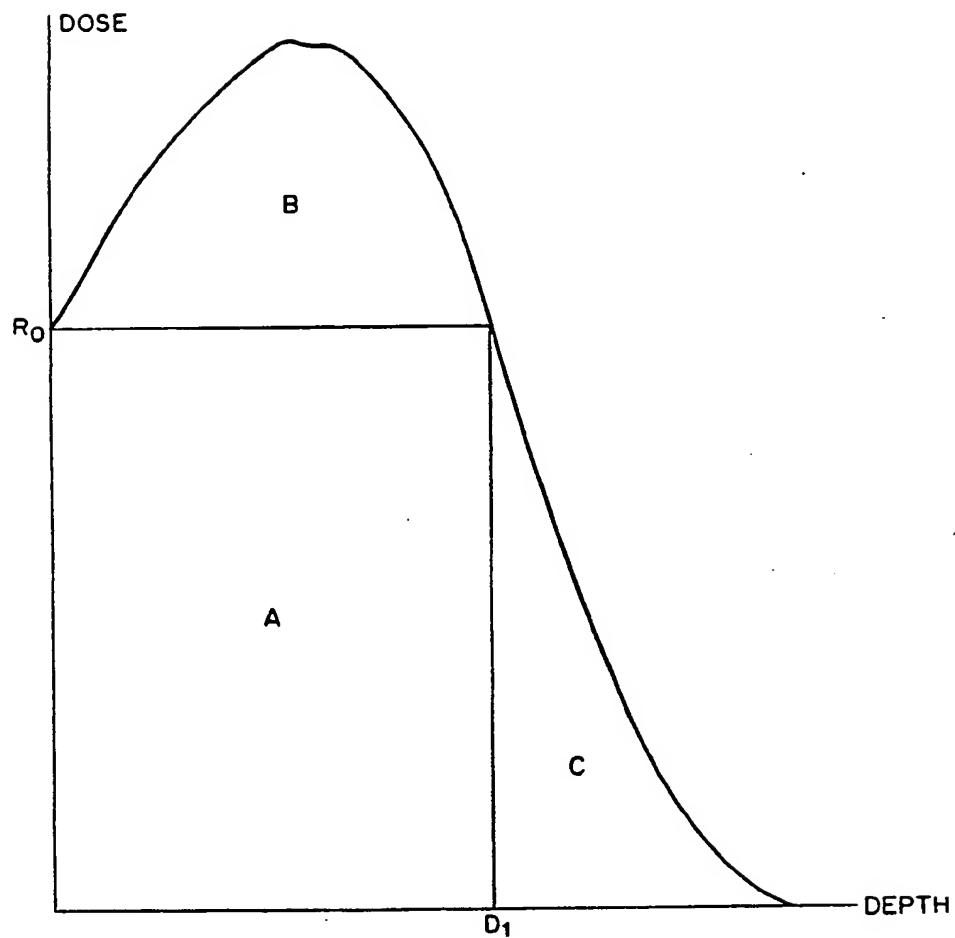


FIG. 6

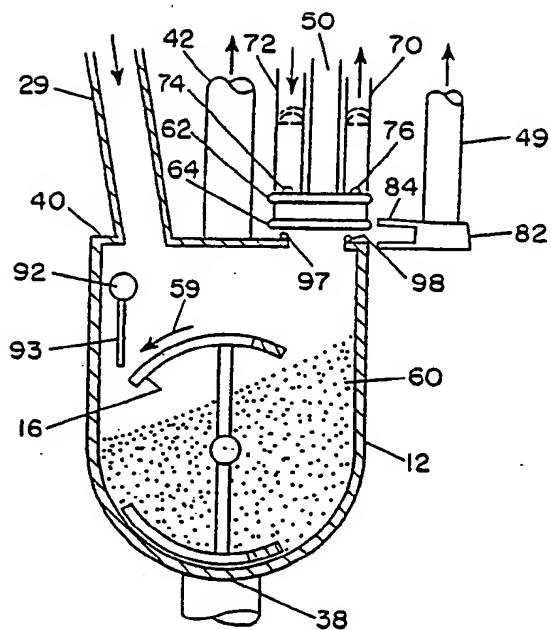


FIG. 7

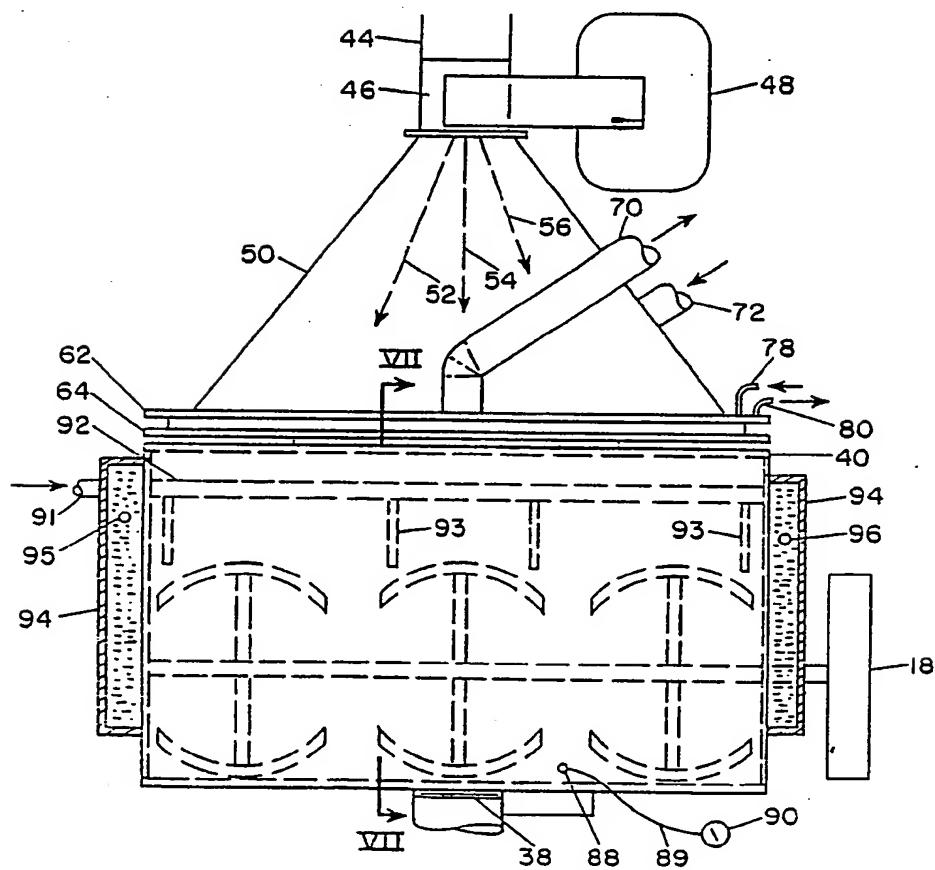


FIG. 8



European Patent  
Office

## EUROPEAN SEARCH REPORT

Application Number

EP 88 10 6777

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl. 4)
X	GB-A-2 119 385 (SHAMROCK CHEMICALS) * Claims 1-19 * -----	1-25	C 08 F 8/50 C 08 F 114/26
TECHNICAL FIELDS SEARCHED (Int. Cl. 4)			
C 08 F			
The present search report has been drawn up for all claims			
Place of search	Date of completion of the search	Examiner	
THE HAGUE	29-11-1988	PERMENTIER W.A.	
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons ..... & : member of the same patent family, corresponding document	